# PATENT SPECIFICATION

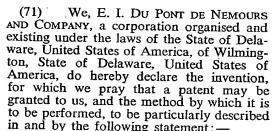
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# (54) OIL COMPOSITIONS



in and by the following statement:—
This invention relates to mineral oil compositions comprising a neutral oil and an ethylene copolymer as a viscosity index

improver for the oil.

It is well known that mineral lubricating oils and functional fluids have a tendency to become thin at elevated temperatures while becoming thick at low temperatures, and thus it is generally necessary to incorporate additives which improve their viscositytemperature relationships. For example, in the case of a crankcase lubricating oil in a cold engine, it is desirable that the oil should not become so thick that it is difficult to 25 start the engine. On the other hand, when the engine is hot, it is necessary that the oil be sufficiently viscous that an oil film is maintained between the moving parts.

The viscosity-temperature relationship of 30 an oil at temperatures in the range of 100° to 210°F. is known as its viscosity index. Thus, additives which retard the tendency of the oil to thin as the temperature is raised from 100° to 210°F. are known as viscosity 35 index improvers. The viscosity index improvers which are most widely used at the present time are polymers of methacrylate esters having long alkyl chains and polyiso-

butylene polymers.

One of the most important considerations from a commercial standpoint in evaluating viscosity index improvers is the thickening power of the additive; that is, the amount of additive necessary to give the desired thickening at 210°F. Since these additives are considerably more expensive than the oil to which they are added, the amount of additive required has a significant effect upon the price of the resulting oil composition. The thickening power of polymeric viscosity index improvers generally increases with increased molecular weight.

One of the problems encountered with polymeric viscosity index improvers is their tendency to thicken mineral oils at low temperatures. In general, lower molecular weight polymers have a reduced tendency to thicken oils at low temperatures and in some cases may even improve their lowtemperature properties.

Another problem frequently encountered with polymeric viscosity index improvers is their lack of shear stability. Shear stability is a measure of the tendency of the oilpolymer composition to become less viscous after prolonged use under high-shear conditions. This loss in the viscosity is believed to be due to a reduction in the molecular weight of the polymer. It is known that low molecular weight polymers tend to be more shear stable than their higher molecular weight counterparts.

Thus the molecular weight of polymeric viscosity index improvers is generally a compromise between a high enough molecular weight to give good thickening power and a low enough molecular weight to give good shear stability and reduced tendency to increase the viscosity of the oil at low temperatures. There is a continuing need for viscosity index improvers which impart to mineral oils an improved combination of viscosity properties including viscosity index, thickening power, low temperature proper-

ties and shear stability.

We have now discovered that certain copolymers of ethylene have outstanding overall properties as viscosity index improvers for mineral oils and according to the present invention we provide an oil composition which comprises a neutral mineral oil base and a viscosity index improver which is an oil-soluble copolymer containing from 25 to 75% by weight of units derived from ethylene and from 75 to 25% by weight of units derived from at least one comonomer selected from: (a) terminally unsaturated straight chain monoolefins having from 3 to





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14 carbon atoms or  $\omega$  - phenyl - 1 - alkenes having 9 or 10 carbon atoms, (b) norbonene, (c) terminally unsaturated non-conjugated diolefins having from 5 to 8 carbon atoms, (d) dicyclopentadiene and (e) 5 - methylene-2 - norbornene, provided that no more than one comonomer from (a) is present in the copolymer, said copolymer having a pendant index of from 4 to 33, an average pendant 10 size not exceeding 10 carbon atoms, an average chain length of from 2700 to 8800 carbon atoms, an inherent viscosity of from 0.7 to 1.8 as measured in a 0.1 weight percent solution in tetrachloroethylene at 30°C., 15 and being present in the oil composition in an amount which improves the viscosity index of the neutral mineral oil base.

The ethylene copolymers used as viscosity index improvers in accordance with this invention impart an outstanding combination of improved viscosity properties to neutral oils containing them. These copolymers have such remarkable thickening powers that they may be used in considerably smaller 25 amounts than the most widely accepted viscosity index improvers in present commercial practice. At the same time, these polymers provide oil-polymer compositions having an overall combination of viscosity 30 index, low temperature viscosity and shear stability properties which is superior to those provided by these same commercial viscosity index improvers.

Preferably, the ethylene copolymer con-35 tains from 25 to 55% by weight of units derived from ethylene and from 75 to 45% by weight of units derived from at least one

of the comonomers specified above.

Suitable terminally unsaturated straight chain monoolefins of 3 to 14 carbon atoms include propylene, 1-butene, 1-pentene, 1-hexene, 1-heptene, 1-octene, 1-nonene, 1-heptene, 1 decene and mixed alkenes of 12 to 14 carbon atoms. Suitable  $\omega$  - phenyl - 1 - alkenes of 9 to 10 carbon atoms are 3 - phenyl - 1propene and 4 - phenyl - 1 - butene. Suitable terminally unsaturated non-conjugated diolefins of 5 to 8 carbon atoms include 1,4pentadiene, 1,4-hexadiene, 1,5-hexadiene, 2-methyl - 1,5 - hexadiene, 1,6 - heptadiene and 1,7 - octadiene.

In general, the copolymers used in accordance with this invention are derived 25 to 75% by weight from ethylene and 75 to 25% from comonomer. In one preferred composition, the copolymers contain from 25 to 75% of units derived from ethylene, from 74 to 20% from propylene and from The most 1 to 20% from 1,4-hexadiene. preferred copolymers are derived from 45 to 60% from ethylene, from 50 to 35% from propylene and from 1 to 10% from 1,4hexadiene. The average chain length of these preferred copolymers is from 4200 to 65 8500 carbon atoms.

The ethylene copolymers useful in the practice of this invention have a pendent index of from 4 to 33. The term "pendent index" is used to indicate the number of pendent groups, such as alkyl, alkenyl, cycloalkenyl and phenylalkyl, per 100 carbon atoms in the backbone of the polymer chain. When the pendent index is greater than 33, poor shear stability may be encountered depending upon the average chain length. 75 Preferably the pendent index is from 10-

The ethylene copolymers have an average pendent size not exceeding 10 carbon atoms. The term "average pendent size" is used to indicate the number of carbon atoms in an average size pendent group. Preferably the pendent size is from 1-6 carbon atoms.

The ethylene copolymers useful in the practice of this invention also have average chain lengths of from 2700 to 8800. The term "average chain length" is used to indicate the average number of carbon atoms in the backbone of the polymer chain as determined by light scattering. It has been found that the average chain length correlates very well with the thickening power of the polymer. When the average chain length of the polymer goes below 2700, the thickening power of the polymer drops At average chain lengths above abruptly. 9000, the shear stabilities of resulting oilpolymer compositions are rather poor.

These ethylene copolymers have inherent viscosities of from 0.7 to 1.8, measured as a 100 0.1% by weight solution of polymer in tetra-chloroethylene at 30°C. The preferred copolymers have inherent viscosities of from 1.1 to 1.7. A definition of inherent viscosity is given in the Journal of Colloid 105 Science, 1, 261—269 (1946). It is expressed

1nNr

wherein "In" is the natural logarithm, "Nr" is the viscosity of the solution relative to 110 the solvent and "c" is the concentration expressed in grams of solute/100 ml. of solvent. Inherent viscosity is indicative of the molecular weight of the polymer. Inherent viscosities of 0.7 to 1.8 correspond to about 115 45,000 to 140,000 weight average molecular weight, as determined by light scattering, while the preferred range of 1.1 to 1.7 corresponds to molecular weights of from 80,000 to 130,000. 120

Optimum performance is achieved with ethylene copolymers within the specified average chain length range which have a relatively narrow molecular weight distribu-Preferably the molecular weight dis- 125 tribution, which is determined by dividing the weight average molecular weight by the

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number average molecular weight, is less than 8.

Suitable copolymers for use in the present invention may be prepared in the manner described in U.S. Patents Nos. 2,799,688, 2,975,159 and 2,933,480. These copolymers are prepared by polymerization in the presence of catalysts generally known as coordination catalysts. These catalysts are made from components of two types, first, compounds of the transition heavy metals of Groups IV, V and VI beginning with titanium, vanadium and chromium, and second, organometallic compounds and hy-15 drides of metals of Groups I, II and III and the free metals of these groups. compounds of the first type are preferably halides, oxyhalides or alcoholates, the preferred metals being titanium and vanadium. The second component is preferably an organometallic compound of lithium, sodium, magnesium or aluminum in which the organic portions are preferably alkyl radicals. In these organometallic compounds the valence of the metal may be partly satisfied by halogen or alkoxyl, provided, of course, that at least one bond connects the metal with an organic radical. Mixtures of two or more compounds of the type de-30 scribed above may often be used to advantage.

As is well known, these catalysts must be used in strict absence of oxygen, water or other material with which they react. For this reason the solvents in which they are used are greatly limited, the preferred one being the saturated aliphatic and hydro-aromatic hydrocarbons and certain nonreactive halogen compounds such as tetra-40 chloroethylene and chlorobenzenes. These compounds conveniently serve as solvents for the polymerization which is usually carried out in a dilute suspension of the catalyst. The polymerization is normally carried out at ordinary temperatures and pressures, although it is usually convenient to allow the temperature to rise spontaneously to about 50° to 60°C. due to the heat of the reaction. Although elevated temperatures and pressures are not required, the rate of the reaction may be increased by the use of increased pressures up to, for instance, about 100 atmospheres or above, or increased temperatures up to about 150°C. Where found desirable, the polymerization may also be carried out at reduced temperatures and pressures. Polymerization conditions are preferably chosen to give a polymer having a narrow molecular weight distribution.

The neutral mineral oil used as the base oil of the composition of this invention may be a lubricating oil, such as the normally used crankcase oils, or a functional fluid such as automotive transmission fluids, and hydraulic fluids. By "neutral mineral oil"

is meant a non-volatile mineral oil which has been refined to remove its acidic and alkaline content, generally by solvent extraction. Solvent extraction may also be used to reduce the paraffin or naphthene content of these oils. The mineral oil may be derived from paraffinic or naphthenic base petroleum, shale oil and the like.

Lubricating oil and transmission fluid base oils are predominantly paraffinic, solvent-refined neutral oils having Saybolt Universal Seconds (SUS) viscosities of from 60 to 220 at 100°F. and viscosity indices of from 80 to 110. Lubricating oils preferably have S.U.S. viscosities of from 90 to 160, while transmission fluids preferably have S.U.S. viscosities of from 60 to 110. Hydraulic fluid base oils are predominantly naphthenic, solvent-refined neutral oils having S.U.S. viscosities not greater than 50 and pour points not above —65°F.

The ethylene copolymer may be incorporated into the base oil by first milling or blending the polymer into a small portion of mineral oil to form a concentrate, which is then blended into the base oil to the desired concentration. Suitable oils for forming the concentrate are paraffinic, naphthenic and mixed neutral oils of 70 to 150 S.U.S. viscosity at 100°F. which are commonly used as solvents or diluents for polymer con-centrates. The solution time may be decreased by preheating the oil to 170° to 210°F. before milling or blending with the polymer. The polymer concentrate may 100 conveniently contain from 5—15% by weight of polymer. The polymer may also be dissolved in the base oil by a solvent transfer technique, whereby the polymer is first dissolved in a volatile solvent, such as 105 carbon tetrachloride, trichloroethylene or nhexene. The solution is then mixed with the base oil and the solvent is removed by distillation.

The effective amount of ethylene co- 110 polymer used in the final oil composition is dependent upon the base oil viscosity. Generally it will be in the range of from 0.5 to 3% by weight and preferably from 1 to 2% by weight.

The oil compositions of this invention may also contain other types of additives usually compounded into neutral oil compositions, such as anti-oxidants, pour point depressants, basic detergents, corrosion inhibitors, 120 rust inhibitors, extreme pressure additives and dyes. The ethylene copolymers used in accordance with this invention are generally compatible with these types of additives.

In order that the invention may be more 125 fully understood, the following Examples are given by way of illustration only. Throughout these Examples, viscosities were measured in accordance with American Society for Testing Materials (ASTM) Test 130

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Method D 445. All percentages are by weight except where the otherwise specified.

EXAMPLE 1

An ethylene/propylene/1,4 - hexadiene

terpolymer was prepared as follows: Tetrachloroethylene solvent (500 ml.) was cooled to 0°C. in a flask equipped with a stirrer, gas delivery tube, thermometer and serum cap. The solvent was saturated with a mixture of ethylene, propylene, nitrogen and hydrogen gases at flow rates of 2.0, 1.5, 0.5 and 0.1 liters per minute respectively. Then 2.9 ml. (0.05 mole) of 1,4-hexadiene was added followed by 5 ml. of a 1.0 diisobutylaluminum solution of chloride in tetrachloroethylene and then 5 ml. of a 0.10 molar solution of vanadium trisacetylacetonate in benzene, each of these being introduced with a hypodermic syringe. The mixture was stirred at 0°C. for 20 minutes and then the catalyst was destroyed

by adding 10 ml. of a 1% solution of 4,41butylidene - bis - (6 - tert. butyl - 3 - methyl phenol) in isopropanol. The reaction mixture was extracted with an equal volume of 5% hydrochloric acid in a high speed mixer and washed with two 500 ml. portions of water. The solvent was allowed to evaporate in an open pan and the polymer residue was vacuum dried for 24 hours at 75°C. and 20 mm. Hg pressure. The yield was 25 g. of ethylene/propylene/1,4 - hexadiene ter-

polymer. The composition of the terpolymer was determined as follows: The propylene content was found to be 45.1% by comparison of the 8.67 to 2.35 micron infrared absorbance ratio with a calibration curve which related this ratio to the propylene content.

This calibration curve was established using radioactive-carbon tagged polymers. The bromine equivalent showed the polymer contained 2.6% unsaturated hexadiene. Applying the 10.36 to 2.35 and 8.67 to 2.34 micron infrared absorbance ratios to a cali-

bration equation gave a total hexadiene content of 3.9%. The inherent viscosity of the terpolymer, measured as a 0.1% solution in tetrachloroethylene at 30°C., was 1.26.

The additional copolymers listed in Tables I and II below were prepared and analyzed in a similar manner.

Additive concentrates of these copolymers were prepared as follows: Twelve gallons (90 lbs.) of a paraffinic, solvent-refined neutral oil of 73 S.U.S. viscosity and a viscosity index of 100 were charged into a 25 gallon capacity Duolater milling apparatus at room temperature. Twelve pounds of copolymer, cut into one-inch cubes, were added. The mixture was milled for about 30 minutes, after which a clear solution resulted, and then for 10 additional minutes. The following tests were carried out using

additive concentrates of the ethylene co-65 polymers listed in Tables I and II.

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Thickening Power

The thickening powers of various ethylene copolymers were determined by measuring the viscosity in centistokes at 100°F. of oil compositions containing a solvent-refined neutral oil having a S.U.S. viscosity of 220 at 100°F. and a viscosity index of 98 as the base oil and sufficient viscosity index improver concentrate to provide a co-polymer concentration of 3%. For comparison, a commercial polyisobutylene polymer, designated "Commercial Polymer A", and a commercial polymethacrylate polymer, designated "Commercial Polymer B" most widely used commercial viscosity index improvers, were also tested.

Viscosity Index

Viscosity index was determined in accordance with ASTM Test Method D 567 by 85 measuring the viscosity at 100°F. of oil compositions containing as the base oil a solvent-refined neutral oil having a S.U.S. viscosity of 107 at 100°F. and a viscosity index 90 of 96, a solvent-refined neutral oil having a S.U.S. viscosity of 130 at 100°F. and a viscosity index of 97 or a blend of the two, and the amount of various ethylene copolymers necessary to give the oil composition a viscosity of 11.5  $\pm$  0.1 centistokes at 210°F. Viscosity index was not affected by the variation in viscosity of the base oil since in all cases the base oil had a viscosity index of 96 to 97. Higher viscosity indices indicate reduced temperature sensitivity of 100 the oil composition.

Weight for Equal Thickening

Weight for equal thickening was determined by measuring the amount of various ethylene copolymers necessary to increase 105 the viscosity of the same base oil as used in the viscosity index determination to 11.5  $\pm$ 0.1 centistokes at 210°F. This amount was then compared with the amount of Commercial Polymer A necessary to accomplish 110 the same result with the same base oil in each case. The amount of Commercial Polymer A was arbitrarily assigned a value of one and the amount of ethylene copolymer was expressed as a fraction thereof. 115

Viscosity at 0°F.

The apparent viscosities at 0°F. of oil compositions containing the same base oil as in the viscosity index test and the amount of various ethylene copolymers necessary to 120 give the the oil composition a viscosity of  $11.5 \pm 0.1$  centistokes at 210°F. were measured using a cold cranking simulator. In this test, a universal motor, run at constant voltage, drives a rotor which is closely 125

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fitted inside a stator. A small sample of the oil composition fills the space between the rotor and stator which are maintained at 0°F. The speed of the rotor is a function of, and is calibrated to determined, the viscosity of the oil composition in poises. Comparisons with Commercial Polymers A and B were run using the same base oils.

Shear Stability

10 The shear stabilities of oil compositions containing the same base oil as in the vis-cosity index test and the amount of various ethylene copolymers necessary to increase the viscosity of the oil composition to 11.5 ± 0.1 centistokes at 210°F, were determined using a 250-w, 10-kc magnetostrictive sonic oscillator in accordance with the technique described in "Proposed Method of

Test for Shear Stability of Polymer-containing Oils", in ASTM Standards, Vol. I, page 1160 (October, 1961). The procedure was modified by shearing a 50 ml. sample for 60 minutes at 100°F, at 0.6 r.f. amps. Shear stability is expressed as the average percent retention of viscosity in centistokes at 210°F. and 100°F. Comparisons with Commercial Polymers A and B were run using the same base oils.

Tables I and II show the improved viscosity properties and the surprising shear 30 stabilities of the ethylene copolymer-containing lubricant compositions of this invention. For comparison, data for oil compositions containing Commercial Polymer A and Commercial Polymer B are given in Table 35

### TABLE I

| Terpolymer   | ,  | Етнул<br>Ва:                                | Visc.                                 | ·                               |                                      |                             |                            |
|--|--|---|---------------------------------------|---------------------------------|--------------------------------------|-----------------------------|----------------------------|
| Weight<br>Ratio  | Average<br>Chain Ti<br>Length                | hickenin<br>Power                           | SUS<br>g at<br>100°F.                 | Viscosity<br>Index              | Wt. for<br>Equal<br>Thickening       | at<br>0°F.,<br>poises       | Shear<br>Stability,<br>%   |
| Ethylene/Propyl<br>Hexadiene   | ene/   |   | · · · · · · · · · · · · · · · · · · · |                                 |                                      |                             |                            |
| 70/28/2<br>62.1/26/11.9<br>59/41/3<br>61.5/35.5/3  | 8500<br>7600 · · ·<br>7400<br>6700           | 880<br>550<br>475                           | 126<br>107<br>131<br>131              | 137<br>141<br>136               | 0.43<br>0.46<br>0.49                 | 7.8<br>7.2                  | 80<br>76<br>83             |
| 41.8/54.7/3.5<br>61.2/36.5/2.3<br>53.1/40.8/6.1<br>50.7/46.8/2.5                             | 6700<br>6400<br>6000<br>6000                 | 448   | 107<br>110<br>107<br>107              | 134<br>140<br>135<br>139<br>142 | 0.48<br>0.52<br>0.51<br>0.60<br>0.56 | 11.5                        | 82<br>81<br>82<br>85<br>85 |
| 54.3/44/1.7<br>57.8/25/17.8<br>55.3/41.0/3.7<br>50.1/46.1/3.8<br>51/45.1/3.9<br>59.7/38/2.3  | 6000<br>5900<br>5600<br>5500<br>5500<br>5300 | 268<br>—<br>317                             | 107<br>107<br>110<br>107              | 144<br>140<br>135<br>144        | 0.56<br>0.60<br>0.56<br>0.62         | 6.0<br>9.1<br>10.2<br>9.6   | 85<br>81<br>               |
| 51.8/45/3.2<br>60.8/31.1/8.1<br>29.3/67.7/3<br>42.5/54.4/3.1<br>55.3/41/3.7<br>42.8/51.6/5.6 | 5200<br>5100<br>5000<br>4900<br>4200<br>3200 | 260<br>———————————————————————————————————— | 122<br>131<br>107<br>107              | 135<br>142<br>135<br>135        | 0.56<br>0.66<br>0.69<br>0.69         | 13.3<br>14.0<br>9.6<br>15.8 | 85<br>90<br>85<br>86       |
| Ethylene/Propyl<br>Dicyclopentad   | ene/   |   |                                       |                                 |                                      |                             |                            |
| 65.9/28.5/5.6  | 7700   | 525   | 131                                   | 138                             | 0.40                                 | <del></del>                 | 64                         |
| Ethylene/Norbor<br>Hexadiene   | rnene/                                       | ·   |                                       |                                 | . •                                  |                             |                            |
| 58.6/38.5/2.9  | 3600   |   | 131                                   | 129                             | 0.78                                 |                             | 97                         |

# TABLE II

# ETHYLENE DIPOLYMERS

| 5  | Dipolyme<br>Weight<br>Ratio                                 | r<br>Average<br>Chain<br>Length                      | Base Oil<br>Visc. SUS<br>at 100°F.     | Viscosity<br>Index                     | Wt. for<br>Equal<br>Thickening               | Visc.<br>at<br>0°F.,<br>poises                     | Shear<br>Stability,<br>%               |  |  |
|----|---|--|--|--|--|--|--|--|--|
|    | Ethylene/Propyler   | ne/  |  |  |  |  |  |  |  |
| 10 | 62/38<br>42/58<br>33/67<br>61/39<br>57/43<br>44/56<br>27/73 | 8200<br>7100<br>6900<br>6600<br>6500<br>5500<br>5400 | 131<br>107<br>107<br>131<br>131<br>107 | 140<br>140<br>135<br>135<br>143<br>141 | 0.39<br>0.40<br>0.62<br>0.44<br>0.48<br>0.59 | 10.2<br>11.6<br>—————————————————————————————————— | 70<br>73<br>83<br>74<br>83<br>84<br>82 |  |  |
| 15 | Ethylene/Hexene   |  |  |  | •  |  |  |  |  |
|    | 43/57<br>32/68  | 4460<br>3200   | 107<br>107                             | 142<br>143                             | 0.56<br>0.72                                 |  | 78<br>81                               |  |  |
|    | Ethylene/Decene   |  |  |  |  |  |  |  |  |
| 20 | 54/46<br>49/51  | 5000<br>3500   | 107<br>107                             | 145<br>144                             | 0.66<br>0.87                                 |  | 70<br>85                               |  |  |
|    | Ethylene/Hexadiene  |  |  |  |  |  |  |  |  |
|    | 69/31   | 5300   | 131                                    | 143                                    | 0.73   |  | 85                                     |  |  |

## TABLE III

# COMPARATIVE DATA

| 25 | Commercial<br>Polymer | Thickening<br>Power             | Base Oil<br>Visc. SUS<br>at 100°F. | Viscosity<br>Index              | Wt. for<br>Equal<br>Thickening | Visc.<br>at<br>0°F.,<br>poises       | Shear<br>Stability,<br>%   |
|----|-----------------------|---------------------------------|------------------------------------|---------------------------------|--------------------------------|--------------------------------------|----------------------------|
| 30 | A<br>A<br>A<br>A      | 142<br>142<br>142<br>142<br>142 | 107<br>110<br>122<br>126<br>131    | 138<br>138<br>138<br>138<br>138 | 1.00<br>1.00<br>1.00<br>1.00   | 14.1<br>14.1<br>14.3<br>14.4<br>14.5 | 77<br>77<br>77<br>77<br>77 |
| ,  | В                     | 157                             | 131                                | 157                             | 1.03                           | 10.7                                 | 62                         |

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EXAMPLE 2
The in-use performance and compatibility of several ethylene copolymers prepared and analyzed as indicated in Example 1 were determined as follows:

Lubricant compositions were prepared eontaining a base oil, an amount of various polymer concentrates, prepared as in Ex-

ample 1, necessary to give a viscosity of 11.5 ± 0.1 centistokes at 210°F., 4% polyaminomonoalkenylsuccinimide, designated Additive X, 1% zinc dialkyldithiophosphate, designated Additive Y, and 2% basic calcium sulfonate, designated Additive Z. The base oil was a blend of refinery components having a S.U.S. viscosity of 107 at 100°F.

and a viscosity index of 97 and containing 20 to 25% by volume aromatics, 10 to 20% by volume olefins, 55 to 65% by volume saturates, 0.21 to 0.25% by weight sulfur and 2.5 to 3 ml. tetraethyl lead per million gallons. The lubricant compositions were tested in a single-cylinder CLR engine operated for 180 hours to determine in-use performance of the polymers and their com-10 patibility with the other additives. The used

lubricant compositions were periodically removed from the engine, topped to remove gasoline dilution, centrifuged to settle out any insoluble matter, and analyzed to determine viscosities at 210° and 100°F.

The following Table shows the changes in viscosity of compounded oils containing various ethylene copolymers compared with the same compounded oils containing Commercial Polymer A.

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TABLE IV PERFORMANCE AND COMPATIBILITY TEST

|     | Weight<br>Ratio<br>Copolym    | Average<br>Chain<br>Length | Other<br>Additives | Temp.,<br>°F. | Visc<br>0    | osity, cer<br>He                      | itistokes<br>ours<br>120 | 180           |
|-----|-------------------------------|----------------------------|--------------------|---------------|--------------|---------------------------------------|--------------------------|---------------|
| F   | Ethylene/Propyle              | ne/                        |                    |               |              | · · · · · · · · · · · · · · · · · · · |                          |               |
| 5   | 57/43                         | 6500                       | X+Y                | 100<br>210    | 69.9<br>11.4 | 63.3<br>10.1                          | 76.4<br>11.0             | 91.5<br>12.4  |
| E   | Ethylene/Propyle<br>Hexadiene | ne/                        |                    |               |              |                                       |                          |               |
| . 5 | 51.8/45/3.2                   | 5200                       | X+Y                | 100<br>210    | 71.9<br>11.5 | 64.9<br>10.2                          | 72.3<br>10.7             | 78.7<br>11.5  |
|     |                               |                            | X+Y+Z              | 100<br>210    | 72.6<br>11.6 | 67.3<br>10.7                          | 72.7<br>11.4             | 108.5<br>18.2 |
|     | Commercial<br>Polymer A       |                            | X+Y                | 100<br>210    | 69.2<br>11.4 | 61.5<br>9.7                           | 89.6<br>12.1             | 144.3<br>20.1 |
|     |                               |                            | X+Y+Z              | 100<br>210    | 71.3<br>11.6 | 65.5<br>10.5                          | 117.4<br>20.2            | 194.9<br>25.9 |

40 From the above Table it can be seen that the viscosity of the compounded oils containing Commercial Polymer thickened considerably after 180 hours, while the compounded oils containing the ethylene copolymers do not exhibit any appreciable thickening. Viscosity increases during prolonged use are generally attributed to oxidation of the oils, producing reactive intermediates. The fact that oils containing the ethylene copolymers do not thicken may be explained by the greater shear stability and thickening power of the copolymers, whereby polymer fragments are not produced which could otherwise react with the 55 reactive oxidation intermediates to form polymer adducts with accompanying vis-

cosity increases. Thus, it has been shown that the novel lubricant compositions of this invention have an outstanding combination of shear stability and viscosity improvement, even when used in exceedingly small amounts, and at low temperatures, and they minimize oil thickening during prolonged use.

WHAT WE CLAIM IS:-65 1. An oil composition which comprises a neutral mineral oil base and a viscosity index improver which is an oil-soluble copolymer containing from 25 to 75% by weight of units derived from ethylene and from 75 to 25% by weight of units derived from at least one comonomer selected from: (a) terminally unsaturated straight chain monoolefins having from 3 to 14 carbon atoms or ω - phenyl - 1 - alkenes having 9 or 10 carbon atoms, (b) norbornene, (c) terminally unsaturated non-conjugated diolefins having from 5 to 8 carbon atoms, (d) dicyclopentadiene, and (e) 5 - methylene - 2 - norbornene, provided that no more

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than one comonomer from (a) is present in the copolymer, said copolymer having a pendant index of from 4 to 33, an average pendant size not exceeding 10 carbon atoms, an average chain length of from 2700 to 8800 carbon atoms, an inherent viscosity of from 0.7 to 1.8 as measured in a 0.1 weight percent solution in tetrachloroethylene at 30°C., and being present in the oil composition in an amount which improves the viscosity index of the neutral mineral oil base.

2. An oil composition according to claim 1 in which the neutral oil is a predominantly paraffinic, solvent-refined, petroleum oil having a S.U.S. viscosity of from 60 to 220 at 100°F. and a viscosity index of from 80 to 110.

3. An oil composition according to claim 2 in which the neutral oil is a lubricating oil having a S.U.S. viscosity of from 90 to 160 at 100°F.

4. An oil composition according to claim 2 in which the neutral oil is a transmission fluid having a S.U.S. viscosity of from 60

to 110 at 100°F.

5. An oil composition according to claim 1 in which the neutral oil is predominantly naphthenic, solvent-refined, petroleum hydraulic fluid having a S.U.S. viscosity of not more than 50 at 100°F. and a pour point of not more than -65°F.

6. An oil composition according to any of claims 1 to 5 in which the ethylene copolymer contains from 25 to 75 weight percent of units derived from ethylene, from 74 to 20 weight percent of units derived from propylene and from 1 to 20 weight percent of units derived from 1,4-hexadiene and has a pendant index of from 10 to 30, an average pendant size of from 1 to 6

carbon atoms and an average chain length of from 4200 to 8500 carbon atoms.

7. An oil composition according to any of claims 1 to 6 in which the ethylene copolymer contains from 45 to 60 weight percent of units derived from ethylene from 50 to 35 weight percent of units derived from propylene and from 1 to 10 weight percent of units derived from 1,4-hexadiene.

8. An oil composition according to any of claims 1 to 5, in which the ethylene copolymer contains from 25 to 55% by weight of units derived from ethylene and from 75 to 45% by weight of units derived from at least one comonomer as specified in claim 1.

9. An oil composition according to any of claims 1 to 8 in which the concentration of ethylene copolymer is from 0.5 to 3 weight percent.

10. An oil composition according to claim 9 in which the concentration of ethylene copolymer is from 1 to 2 weight percent.

11. A viscosity index improver additive concentrate which is a modification of the oil composition as claimed in any of claims 1 to 8 in which the concentration of ethylene copolymer is from 5 to 15 weight percent.

12. An oil composition according to claim 1 substantially as herein described in either of the Examples.

13. An additive concentrate according to claim 11 substantially as herein described in 75 Example 1.

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